



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

KEI MORIMOTO ET AL

SERIAL NO. 09/630,998      GROUP ART UNIT: 1711

EXAMINER: WOODWARD, ANA LUCRECIA

FOR: POLYAMIDE RESIN COMPOSITION HAVING IMPROVED WEATHERING  
RESISTANCE AND ITS MOLDED PRODUCTS

DECLARATION UNDER 37 C.F.R. 1.132

HONORABLE COMMISSIONER OF PATENTS & TRADEMARKS

WASHINGTON, D.C. 20231

SIR:

Now comes Masaki HIRONO a citizen of Japan, and a resident of c/o Mitsubishi Engineering-Plastics Corporation, Technical Center, 6-2, Higashi-Yahata 5-chome, Hiratsuka-shi, Kanagawa-ken, Japan, who declares and says that:

1. I graduated from the post-graduate course of Applied Chemistry, Kumamoto University, in March, 1993.

2. I have was an employee worked as a plastic R&D assistance of Mitsubishi Gas Chemical Company Inc. for 1993-1994, and has been an employee of Mitsubishi Engineering-Plastics Corporation since 1994 and have worked as a plastic engineer.

3. I am one of inventors of U.S. Patent Application, Serial No. 09/630,998.

4. I have read the Office Action dated March 20, 2003, have understood the Examiner's rejection of the invention claimed in the above application. Then, under my control, the following Experiments were conducted.

(1) Experiments 1-2 (within the scope of the present invention)

The weighed quantities of the component materials which are shown in Table, were mixed by a tumbler, melted and then kneaded at 270°C by a vent type extruder, extruded into a string, cooled in a water bath, cut, dried and then pelletized. To the obtained products, the following evaluation tests were conducted. The accelerated deterioration test was carried out by Super UV test using a square mirror-surface plate as a test piece for 300 hours. The results of evaluation tests are shown in Table 1.

(2) Experiment 3 (corresponding to Example 2 of EP 0839862)

The weighed quantities of the component materials which are shown in Table 1 and correspond to those of Example 2 of EP 0839862, were mixed by a tumbler, melted and then kneaded at 270°C by a vent type extruder, extruded into a string, cooled in a water bath, cut, dried and then pelletized. To the obtained products, the following evaluation tests were conducted. The accelerated deterioration test was carried out

by Super UV test using a square mirror-surface plate as a test piece for 300 hours. The results of evaluation tests are shown in Table 1.

(3) Experiment 4 (corresponding to Example 3 of EP 0839862)

The weighed quantities of the component materials which are shown in Table 1 and correspond to those of Example 3 of EP 0839862, were mixed by a tumbler, melted and then kneaded at 270°C by a vent type extruder, extruded into a string, cooled in a water bath, cut, dried and then pelletized. To the obtained products, the following evaluation tests were conducted. The accelerated deterioration test was carried out by Super UV test using a square mirror-surface plate as a test piece for 300 hours. The results of evaluation tests are shown in Table 1.

The used materials in Experiments 1-4 and evaluation methods are set forth below.

<Materials>

- (1) Polyamide 6 resin: NOVAMID 100 7J (trade name) produced by Mitsubishi Engineering-Plastics Corporation; relative viscosity: 2.2.
- (2) Polyamide 6 resin (Nylon 6): 1011FB (trade name) produced by Ube Industries, Ltd.
- (3) Aromatic polyamide resin: aromatic polyamide A obtained in the following method.

Adipic acid was melted by heating in a reaction vessel under a nitrogen atmosphere. To the molten dicarboxylic acid,

a xylylenediamine mixture consisting of 30 mol% of paraxylylenediamine and 70 mol% of methaxylylenediamine was added dropwise, and the solution was stirred while maintaining the reaction temperature so that it would always remain higher than the melting point of the product. After completion of the dropwise addition, stirring and reaction were continued until the prescribed viscosity was reached, at which point the reaction product was taken out of the reaction vessel, cooled with water and pelletized. The thus obtained aromatic polyamide A had a melting point of 258°C, a crystallization temperature of 206°C and a relative viscosity (measured in a 96% sulfuric acid solution at a concentration of 1 g/100 ml) of 2.08.

- (4) Polyamide MXD6 (Nylon MXD6): 6007 (trade name) produced by Mitsubishi Gas Chemical Company, Inc.
- (5) Copper iodide
- (6) Potassium iodide
- (7) Phenolic antioxidant: N,N'-hexamethylenebis(3,5-di-t-butyl-4-hydroxy-hydrocinnamamide, Irganox 1098 (trade name) produced by Ciba Geigy (Japan) Ltd.
- (8) Phosphoric antioxidant: bis(2,6-di-t-butyl-4-methylphenyl)pentaerythritol diphosphite), ADEKA SUTABU PEP-36, produced by Asahi Denka Kogyo K.K.

#### <Evaluation methods>

##### (1) Moldability

A 100 × 150 × 50 mm box with a thickness of 2 mm was molded under the conditions of: resin temperature = 275°C;

mold temperature = 90°C; injection pressure = 500 kgf/cm<sup>2</sup>;  
injection speed = 50 mm/sec; dwelling pressure = 300 kgf/cm<sup>2</sup>;  
injection pressure dwelling time = 12 sec; cooling time = 20  
sec, and releasability in the molding operation and visual  
appearance of the molded article (box) were evaluated into the  
following three-grade.

○: Releasability was good, and there was noted little surface  
roughening.

△: Releasability was rather bad, and surface roughening was  
observed.

×: Releasability was bad, and surface roughening was  
excessive.

## (2) Accelerated deterioration test

A 70 mm × 70 mm square mirror-surface or embossing  
texture plate with a thickness of 3 mm was molded under the  
conditions of: resin temperature = 275°C; mold temperature =  
90°C; injection pressure = 500 kgf/cm<sup>2</sup>; injection speed = 30  
mm/sec; dwelling pressure = 200 kgf/cm<sup>2</sup>; injection pressure  
dwelling time = 12 sec; cooling time = 20 sec, and it was  
subjected to the following three accelerated deterioration  
tests. Weathering resistance was evaluated by the changes of  
glossiness and color difference after the test. The testing  
conditions and the method of determination of glossiness are  
shown below.

### 1) Super UV test

(Testing conditions)

Tester: Eye super UV (trade name)

Test piece: square mirror-surface plate (molded by a  
#3000 mirror polished mold)

Light intensity: 50 mW/cm<sup>2</sup> (295-450 nm)

Humidity: not less than 90% in RH

Temperature: 63±3°C

Test time: 200 or 300 hours

## 2) Determination of glossiness

Glossiness was determined according to the method of JIS Z-8741 using NIPPON DENSHOKU GLOSS METER VG2000. Measurement was made at an angle of 60 degrees. The greater the measured value, the closer is the molded article (plate) surface to the mirror-surface state.

## (3) Surface observation test (Surface condition)

The square plate after the accelerated deterioration test obtained according to the method of (2) described above was observed under a stereo microscope, and the surface condition was evaluated into the following 4-grade.

◎: Almost no surface roughening was noted.

○: Slight surface roughening was observed.

△: Surface roughening was seen.

×: Excessive surface roughening was seen.

Table 1

Experiment	1	2	3	4
<u>Composition (parts by weight)</u>				
Polyamide 6 resin (NOVAMID 100 7J)	70	70	-	-
Polyamide 6 resin (1011FB)	-	-	30	70
Aromatic polyamide resin	30	30	-	-
Polyamide MXD6 (6007)	-	-	70	30
Copper iodide	-	0.02	0.02	0.02
Potassium iodide	-	0.4	0.4	0.4
Phenolic antioxidant (Irganox 1098)	-	0.5	0.5	0.5
Phosphoric antioxidant (ADEKA SUTABU PEP-36)	-	0.5	0.5	0.5
<u>Evaluation</u>				
Moldability	○	○	△	○
Accelerated deterioration test (Glossiness at 60°)				
·Initial	112	114	119	115
·After 300 hours irradiation	49	70	18	36
Surface condition	○	◎	×	△

(4) Remarks

In comparison with Experiment 2 (present invention) and Experiments 3-4 (Examples 2 and 3 of EP 0839862), only the used polyamide resin were different and the polyamide resin composition of Experiment 2 is superior to the compositions of Experiments 3 and 4 in the moldability, accelerated deterioration test and surface condition.

In Experiment 1 (present invention), the polyamide resin composition did not contain additives (copper iodide, potassium iodide, phenolic antioxidant (Irganox 1098) and phosphoric antioxidant (ADEKA SUTABU PEP-36)) which were contained in the compositions of Experiments 3 and 4. However, even though not containing such additives in Experiment 1, the polyamide resin composition in Experiment 1 is superior to the Experiments 3 and 4 in the accelerated deterioration test and surface condition.

Therefore, in both cases of containing and not containing the additives, the polyamide resin composition of the present invention is superior to the composition of EP 0839862 in the accelerated deterioration test and surface condition.



5. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

6. Further, deponent saith not.

Date: October 15th, 2003

M. Hirono

Masaki HIRONO